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THERMODYNAMIC PROPERTIES OF
POLYMETHYL METHACRYLATE AND
METHYL METHACRYLATE

NOL

17 JANUARY 1962

UNITED STATES NAVAL ORDNANCE LABORATORY, WHITE OAK, MARYLAND

NOLTR 62-98

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THERMODYNAMIC PROPERTIES OF POLYMETHYL METHACRYLATE
AND METHYL METHACRYLATE

Prepared by:
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ABSTRACT: Lack of information on the fundamental thermodynamic properties of polymethyl methacrylate and methyl methacrylate has prompted a study of the available thermodynamic parameters of these materials. From published data the entropy, enthalpy, and Gibbs free energy values of polymethyl methacrylate have been calculated over the temperature range 0° to 260°K. The thermodynamic function, C_p/T vs. T , has been calculated and exhibits a maximum value between 70° and 100°K. An approximation is made of the number of vibrating units per repeating unit of polymethyl methacrylate at 260°K. The difference $C_p - C_v$ is calculated at 260°K and found to be 0.014 cal/deg. gm. It is recommended that additional low temperature specific heat measurements be made on polymers and monomers.

PUBLISHED AUGUST 1962

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NOLTR 62-98

17 January 1962

THERMODYNAMIC PROPERTIES OF POLYMETHYL METHACRYLATE AND METHYL
METHACRYLATE

This report contains the results of an analysis of the thermodynamic properties of polymethyl methacrylate and methyl methacrylate. The calculated values of the thermodynamic functions, entropy, enthalpy and Gibbs free energy, are the first published values of these functions. Data of the type considered in this report are fundamental properties of the materials considered and are a contribution toward a basic understanding of these materials. This investigation was derived from work done under project N10-a-1-56 as part of a general polymer investigation.

W. D. COLEMAN
Captain, USN
Commander

Albert Lightbody
ALBERT LIGHTBODY
By direction

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INTRODUCTION

1. A fundamental approach toward an understanding of the structure and internal motions of a polymer is to investigate the temperature dependence and magnitude of the specific heat, C_p , and of the related thermodynamic functions over a temperature range extending down to absolute zero. Changes in C_p and in the related functions entropy, enthalpy, and free energy are indicative of changes in the structure and internal motions of a polymer due to crystallization, glass transitions, and melting. In addition, C_p measurements at low temperatures are useful in testing the validity of the various theoretical equations for the heat capacity of linear polymers, such as those advanced by Tarassov (ref. (a)) and by Stockmayer and Hecht (ref. (b)).

2. Additional thermodynamic data on bulk polymers are needed to elucidate the nature of the glassy and crystalline states and to explain the magnitude of the energy changes which occur when polymers are heated or cooled, crystallized or melted. This report, which is the fourth in a series (refs. (c), (d), (e)), presents the calculated values of the entropy, enthalpy, and Gibbs free energy for polymethyl methacrylate (PMMA) over the temperature range 0° to 260°K . These calculations are based upon published specific heat data (refs. (f) and (g)), over the range 16° to 260°K . Also presented are the values of the entropy, enthalpy, and Gibbs free energy for methyl methacrylate monomer (MMA) over the temperature range 0° to 210°K . The thermodynamic function C_p/T vs. T , the difference $C_p - C_v$, and the number of classically vibrating units per repeating unit of PMMA are also calculated and discussed.

THE SPECIFIC HEAT OF POLYMETHYL METHACRYLATE

3. Specific heat of polymethyl methacrylate is of particular interest because of the presence of pendent $-\text{CH}_3$ and $-\text{CO}-\text{OCH}_3$ groups along the main chain. It would be expected that the ester methyl group in PMMA is more capable of different types of motion than is the phenyl group in polystyrene (PS) and that the main chain methyl group in PMMA will also contribute to the specific heat. Thus, while the molecular weight per repeating units of PMMA and PS are very similar it would be expected that the differences in the vibrational spectrum produced by the various pendent groups would be reflected by differences in the temperature dependence of the specific heats.

4. Specific heat of polymethyl methacrylate has been determined by Sochava (ref. (g)) between 16° and 60°K and by Sochava and Trapeznikova (ref. (f)), between 60° and 260°K . Sochava and Trapeznikova (ref. (f)) have also determined the specific heat of methyl methacrylate between 60° and 210°K , and Erdos, Jager and Pouchly (ref. (h)) have determined its specific heat between 293° and 323°K . The

experimental values are given in Tables 1 and 2 and are presented graphically in Figure 1.

5. Specific heat of polymethyl methacrylate between 0° and 16°K was obtained by extrapolating to absolute zero a plot of C_p/T vs. T^2 of the experimental data between 16° and 30°K. Specific heat of methyl methacrylate between 0° and 60° was estimated by means of a Debye function (ref. (i)) with eight degrees of freedom and θ_D of 151°K.

6. A plot of the observed low temperature C_p of PMMA and PS is shown in Figure 2. It will be noted that the C_p of PMMA increases in a linear manner between 16° and 60°K while the C_p of PS exhibits a non-linear temperature dependence. At low temperatures amorphous linear polymers such as PMMA and PS should exhibit a linear increase in C_p providing that no interactions occur between polymer chains and that the observed C_p is due to simple transverse vibrations. It thus appears that at low temperatures PS has an additional C_p superimposed upon a linear C_p . Trapeznikova and Feofanova (ref. (j)) have concluded that this additional C_p is due to the rotation of the phenyl group in PS.

7. While the specific heat data do appear to indicate that more than simple vibrations of the main chain PS are taking place, these data do not indicate just what groups or types of vibrations are involved. In the case of polymethyl methacrylate Sinnott (ref. (k)) has concluded that the ester and main chain methyl groups progressively exhibit increased motion with increasing temperature. Motion of these groups would contribute to the specific heat. Sochava (ref. (g)) has recently pointed out that the higher specific heat of polymethyl methacrylate at temperatures above 50°K, when compared with polystyrene, is due to the torsional vibrations of the methyl groups.

8. Warfield and Petree (ref. (l)) have recently shown that the model of Stockmayer and Hecht (ref. (b)) using the analytical procedure developed by Starkweather (ref. (m)) can be applied to polymethyl methacrylate and to polystyrene and that a number of parameters indicative of the structure of the polymer can be calculated from C_p data.

ENTROPY, ENTHALPY AND GIBBS FREE ENERGY CALCULATIONS

9. From C_p data, values of entropy, enthalpy, and Gibbs free energy of polymethyl methacrylate and methyl methacrylate have been calculated at ten degree increments by numerical integration and are presented in Tables 1 and 2 and in Figure 3. The values were obtained by evaluating the thermodynamic relations

$$S_T - S_{0^\circ K} = \int_0^T \frac{C_p dt}{T} \quad (1)$$

$$H_T - H_{0^\circ K} = \int_0^T C_p dt \quad (2)$$

$$(F_T - F_{0^\circ K}) = (H_T - H_{0^\circ K}) - T (S_T - S_{0^\circ K}) \quad (3)$$

where $(S_T - S_{0^\circ K})$, $(H_T - H_{0^\circ K})$, and $(F_T - F_{0^\circ K})$ are the entropy, enthalpy, and Gibbs free energy relative to absolute zero.

10. Since polymethyl methacrylate is an amorphous polymer it must be assumed that there will be residual entropy at absolute zero. While there is some evidence that this residual entropy will be small (ref. (n)), its magnitude is unknown.

11. At 260°K the entropy of polymethyl methacrylate was found to be 0.3375 cal/gm. deg. or 33.75 cal/mole deg. Entropy of methyl methacrylate at 210°K was found to be 0.3788 cal/gm. deg. or 37.88 cal/mole deg. and at 210°K the entropy of polymerization was found by difference to be 9.63 cal/mole deg. At 260°K the enthalpy of polymethyl methacrylate was found to be 40.11 cal/gm. and that of methyl methacrylate at 210°K was 37.50 cal/gm. The Gibbs free energy of polymethyl methacrylate at 260°K was found to be -45.95 cal/gm. and for methyl methacrylate at 210°K -40.15 cal/gm.

12. The free energy of polymerization, ΔF , is calculated at 210°K by means of Equation (4).

$$\Delta F = \Delta H - T\Delta S \quad (4)$$

Where ΔH is the heat of polymerization of MMA, 13.8 Kcal/mole (ref. (o)) and ΔS is the entropy of polymerization, -9.6 cal/mole deg. The free energy of polymerization is thus found to be -11.8 Kcal/mole at 210°K. This ΔF value may be slightly high since the ΔH was determined at 300°K. The corresponding value for styrene is -9.4 Kcal/mole at 300°K. These values can be compared with the ΔF value of -14.3 Kcal/mole which has been calculated by Dainton, et al. (ref. (p)) for the polymerization of 3,3 bis(chloromethyl) oxacyclobutane.

13. Plots of the thermodynamic functions vs. temperature for the polymer are shown in Figure 3. These functions change with temperature in a manner very similar to those of polystyrene (ref. (c)) and polyethylene (ref. (d)).

THE FUNCTION C_p/T VS. T FOR POLYMETHYL METHACRYLATE

14. Smith and Dole (ref. (q)) have pointed out that the function C_p/T vs. T is a constant if the specific heat rises linearly with T from a zero value at absolute zero. The function C_p/T measures the rate at which the entropy increases with temperature or

$$\left[\frac{dS}{dT} \right]_P = \frac{C_p}{T} \quad (5)$$

Dole and Wunderlich (ref. (r)) have presented data for the magnitude and temperature dependence of this function for a number of polymers and long chain hydrocarbons. For many of the hydrocarbons a maximum value of the function is observed at 60° to 70°K which, according to Dole and Wunderlich (ref. (r)), is due to one or more modes of vibration having the same frequency dominating at this temperature range. Warfield and Petree have shown that polystyrene (ref. (c)), polyethylene (ref. (d)), polyvinyl alcohol (ref. (e)), and polytetrafluoroethylene (ref. (d)) exhibit maxima at temperatures below 100°K.

15. A plot of C_p/T vs. T for polymethyl methacrylate is shown in Figure 4. Data for polystyrene are included for purposes of comparison. It will be noted that polymethyl methacrylate exhibits a maximum value of this function between 70° and 100°K and that at higher temperatures the magnitude of the function decreases. These maximum values of the C_p/T vs. T function are probably due to interactions between chains which can best be described by a three-dimensional Debye continuum of acoustic frequencies (ref. (i)).

VIBRATING UNITS PER REPEATING UNIT OR POLYMETHYL METHACRYLATE

16. Based upon a number of assumptions, Dole (ref. (s)) was able to calculate the heat capacity per vibrating unit of polyethylene. Employing the same assumptions it is possible to estimate the number of classically vibrating units at 260°K in the PMMA repeating unit $-C(CH_3)(COOCH_3)CH_2-$. Three assumptions are employed. The first is that the force constant of the C-H bond is so great that the methyl and methylene groups will vibrate as a single unit; the second is that every group in the chain can vibrate harmonically with two degrees of freedom along mutually perpendicular axes transverse to the chain direction, and thirdly that longitudinal or stretching vibrations are negligible at 260°K. If each vibrating unit vibrates with two degrees of freedom, the expected specific heat would be 2R or 3.97 cal/deg. mole per vibrating unit. However, these considerations apply to C_v , not C_p .

17. The number of vibrating units per repeating unit is found by dividing C_v , the heat capacity at constant volume, by 2R. C_v can be calculated by means of Equation (6).

$$C_v = C_p \left[\frac{1}{1 + \frac{TV^2d^2}{C_p J}} \right] \quad (6)$$

Where C_p is the observed heat capacity of PMMA at 260°K, 0.267 cal/deg. gm or 26.7 cal/mole deg.; V is the sound velocity, 2320 m/sec (ref. (t)); d is the cubic coefficient of expansion, 2.06×10^{-4} per deg (ref. (u)); and J is the mechanical equivalent of heat, 4.184×10^{-7} erg/cal. C_v is found to be 0.253 cal/deg. gm. or 25.3 cal/mole deg. and $C_p - C_v$ is 0.014 Cal/gm. deg. Based upon these values the number of vibrating units per repeating unit of PMMA at 260°K is 6.38.

CONCLUSIONS

18. Considerations of data presented in this report have led to the following conclusions:

a. The low temperature specific heat of polymethyl methacrylate and polystyrene exhibit small but significant differences which can be attributed to the motion of pendent side groups.

b. $C_p - C_v$ for polymethyl methacrylate at 260°K is 0.014 cal/deg. gm.

c. The number of vibrating units per repeating unit of PMMA is calculated to be 6.38.

RECOMMENDATIONS

19. It is recommended that calorimetric measurements be made to obtain low temperature specific heats of many of the common monomers and polymers. Data of this type would permit calculation of the entropy, enthalpy, and free energy of these systems and the changes in these values due to polymerization. A complete thermodynamic investigation of the factors that make up the observed specific heat over a broad temperature range would be of great importance toward a more complete understanding of the structure and internal motion of a polymer.

TABLE 1

SPECIFIC HEAT, ENTROPY, ENTHALPY,
AND GIBBS FREE ENERGY OF METHYL METHACRYLATE

Temperature °K	Specific Heat, $C_p^{(1)}$ cal/deg. gm.	$S_T - S_0^{\circ K}$ cal/deg. gm.	$H_T - H_0^{\circ K}$ cal/gm.	$-(F_T - F_0^{\circ K})$ cal/gm.
10	(0.004)	0.0021	0.013	0.002
20	(0.027)	0.0153	0.133	0.097
30	(0.059)	0.0372	0.562	0.368
40	(0.084)	0.0616	1.288	0.868
50	(0.103)	0.0856	2.242	1.610
60	0.118	0.1079	3.352	2.582
70	0.142	0.1298	4.662	3.775
80	0.157	0.1512	6.162	5.178
90	0.170	0.1717	7.802	6.792
100	0.181	0.1912	9.552	8.162
110	0.191	0.2097	11.40	10.62
120	0.204	0.2276	13.37	12.80
130	0.217	0.2452	15.48	15.17
140	0.230	0.2624	17.71	17.71
150	0.244	0.2793	20.08	20.42
160	0.256	0.2960	22.58	23.30
170	0.269	0.3124	25.20	26.34
180	0.285	0.3287	27.97	29.55
190	0.305	0.3451	30.92	32.92
200	0.328	0.3617	34.08	36.45
210	0.358	0.3788	37.50	40.15

(1) Data of Sochava (60° to 210°K)

TABLE 2

SPECIFIC HEAT, ENTROPY, ENTHALPY,
AND GIBBS FREE ENERGY OF POLYMETHYL METHACRYLATE

Temperature °K	Specific Heat, $C_p^{(1)}$ cal/deg. gm.	$S_T - S_0^{\circ K}$ cal/deg. gm.	$H_T - H_0^{\circ K}$ cal/gm.	$-(F_T - F_0^{\circ K})$ cal/gm.
10	(0.010)	0.0112	0.070	0.010
20	0.019	0.0246	0.190	0.179
30	0.035	0.0384	0.460	0.500
40	0.051	0.0529	0.890	0.962
50	0.066	0.0675	1.470	1.568
60	0.083	0.0823	2.210	2.316
70	0.098	0.0977	3.130	3.220
80	0.112	0.1127	4.180	4.272
90	0.126	0.1277	5.380	5.474
100	0.139	0.1425	6.710	6.828
110	0.149	0.1569	8.150	8.324
120	0.158	0.1708	9.680	9.962
130	0.166	0.1843	11.30	11.74
140	0.176	0.1975	13.02	13.64
150	0.186	0.2104	14.83	15.68
160	0.194	0.2231	16.73	17.85
170	0.202	0.2356	18.72	20.15
180	0.210	0.2478	20.79	22.58
190	0.218	0.2597	22.93	25.11
200	0.226	0.2709	25.15	27.68
210	0.235	0.2825	27.47	30.44
220	0.244	0.2939	29.86	33.33
230	0.250	0.3051	32.34	36.31
240	0.255	0.3161	34.87	39.41
250	0.262	0.3269	37.46	42.63
260	0.267	0.3375	40.11	45.95

(1) Data of Sochava (16° to 260°K)

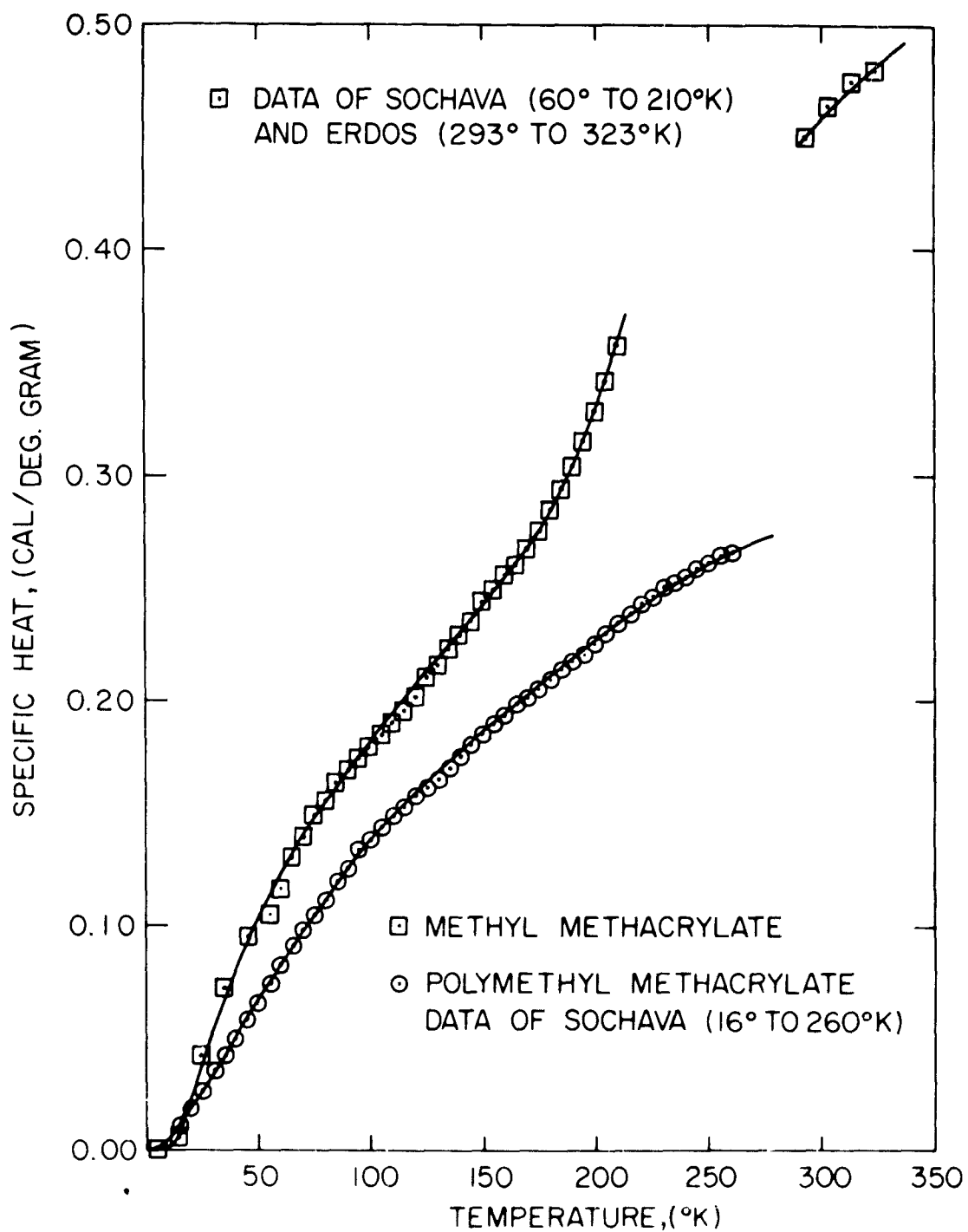


FIG. 1 SPECIFIC HEAT OF METHYL METHACRYLATE AND POLYMETHYL METHACRYLATE.

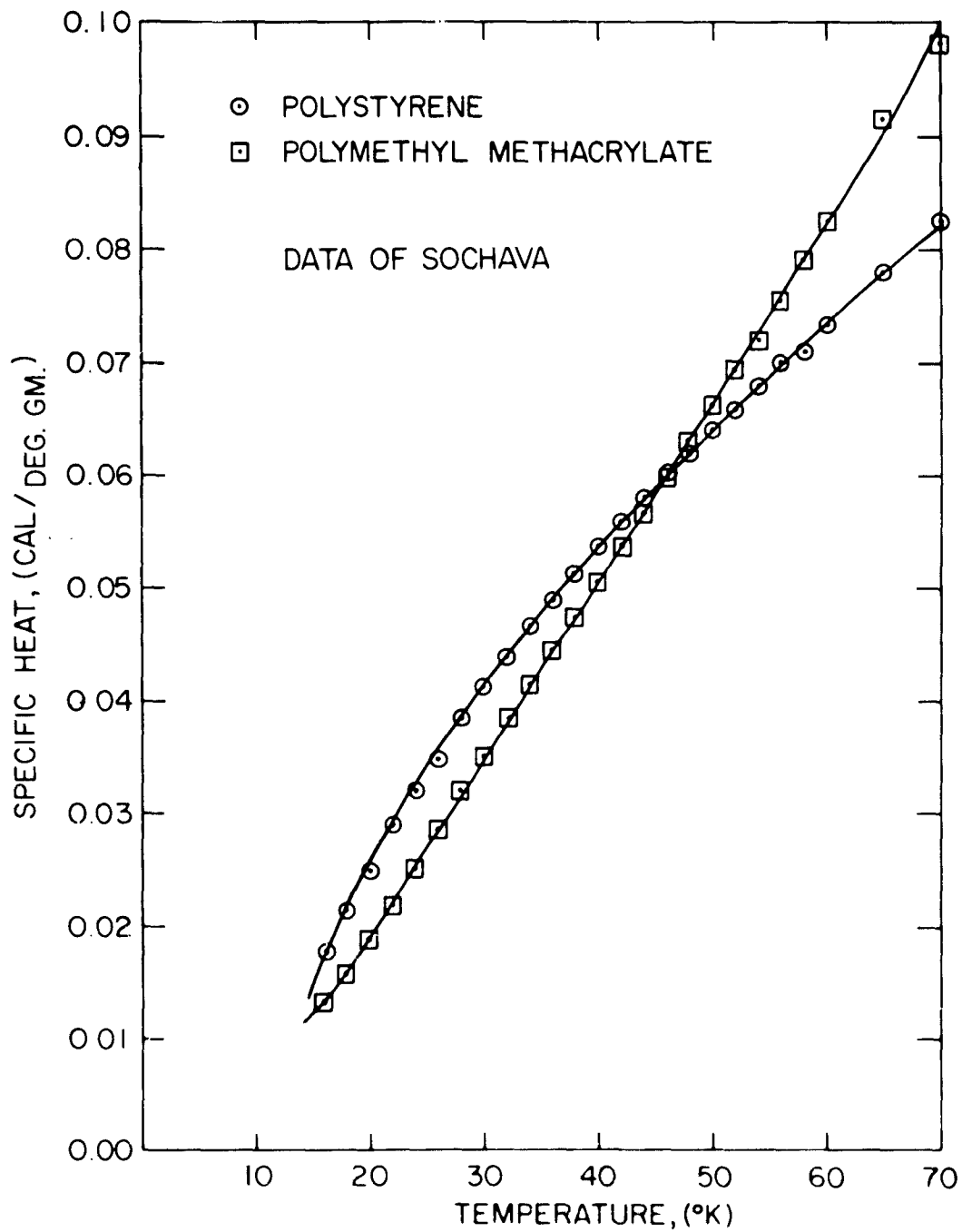


FIG. 2 LOW TEMPERATURE SPECIFIC HEAT OF
POLYSTYRENE AND POLYMETHYL METHACRYLATE.

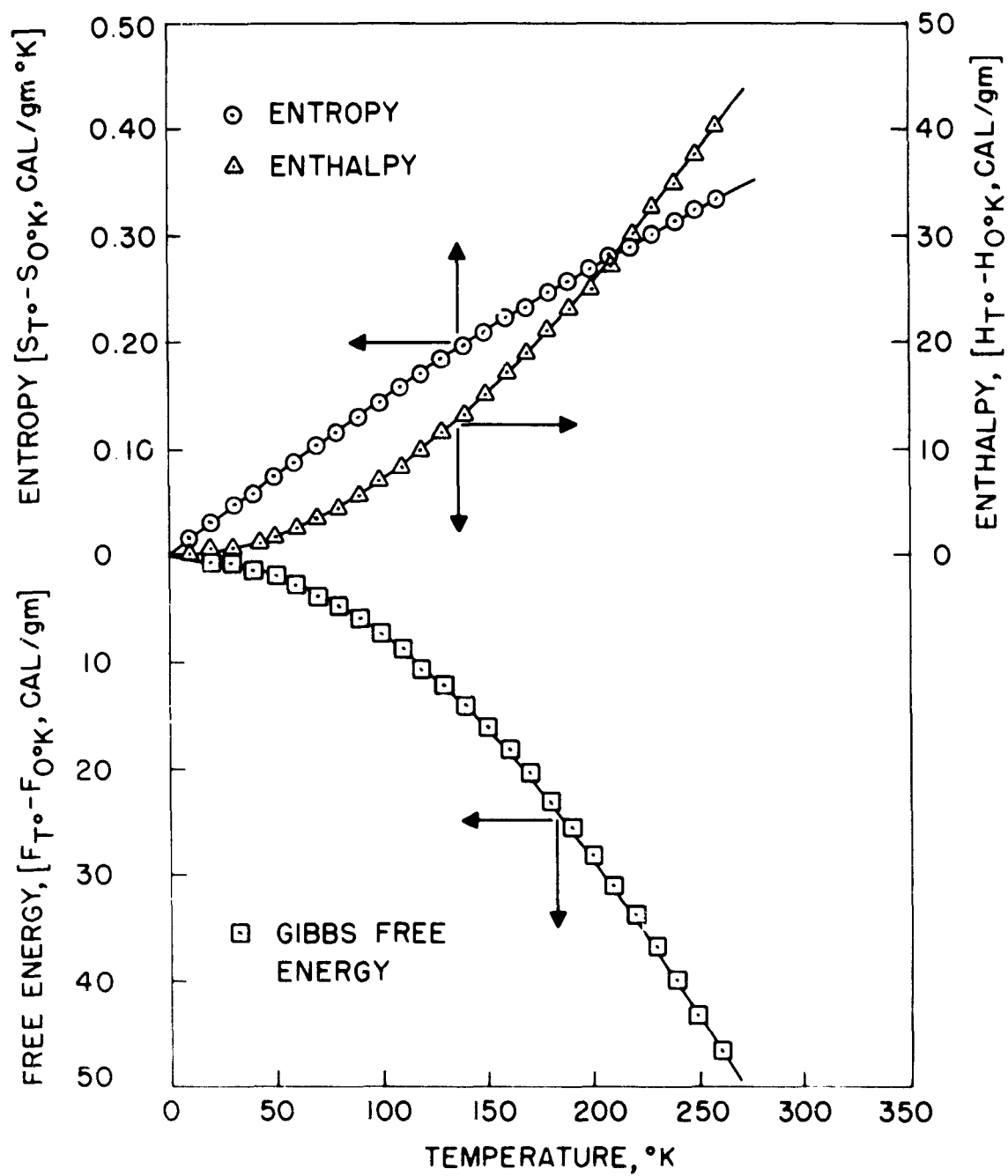


FIG 3 THERMODYNAMIC FUNCTIONS OF
POLYMETHYL METHACRYLATE

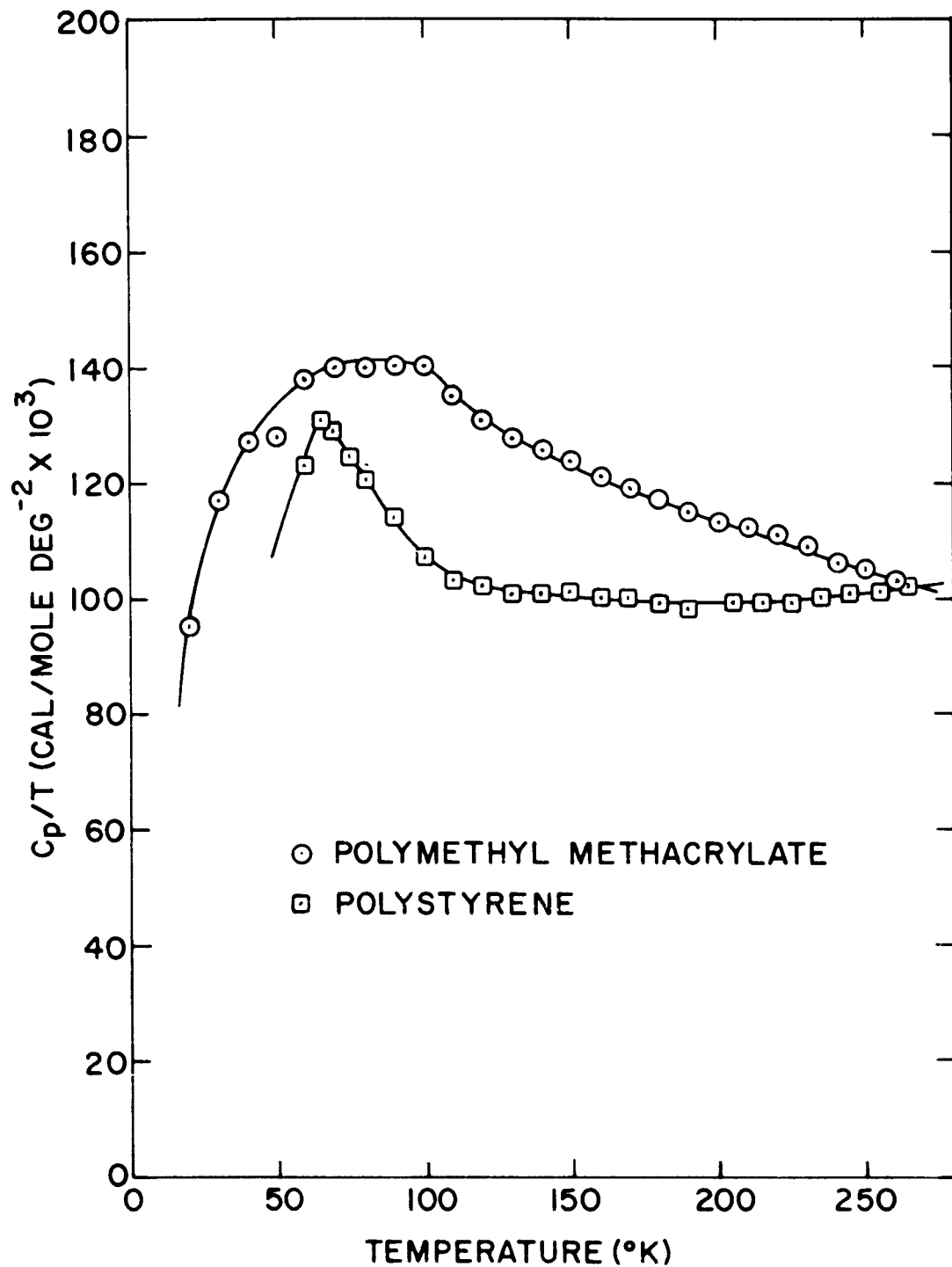


FIG. 4 SPECIFIC HEAT FUNCTION C_p/T VS T FOR POLYMETHYL METHACRYLATE AND POLYSTYRENE

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	DESCRIPTORS	CODES	SECURITY CLASSIFICATION AND CODE COUNT	DESCRIPTORS	CODES
SOURCE	NOI technical report	NOLTR		Unclassified - 13	U013
REPORT NUMBER	62-94	620094	CIRCULATION LIMITATION		
REPORT DATE	17 Jan. 1962	Q162	CIRCULATION LIMITATION OR BIBLIOGRAPHIC		
			BIBLIOGRAPHIC (SUPPL., VOL., ETC.)		

SUBJECT ANALYSIS OF REPORT

	DESCRIPTORS	CODES	DESCRIPTORS	CODES
Polymethyl	POLM		Plastics	PLAS
Methacrylate	METC			
Methyl	METY			
Thermodynamics	THED			
Polymers	POLY			
Monomers	MONM			
Entropy	ENTP			
Enthalpy	ENTH			
Gibbs	GBGS			
Energy	ENER			
Calculation	COMA			
Vibration	VIBP			

Naval Ordnance Laboratory, White Oak, Md.
(NOL technical report 62-98)
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METHACRYLATE AND METHYL METHACRYLATE (U) by
R.W. Warfield and M.C. Petree. 17 Jan. 1962.
7p. charts, tables. Project N10-a-1-56.
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Abstract card is unclassified

1. Plastics
Thermodynamics
2. Polymethyl
methacrylate
3. Methyl meth-
acrylate
- I. Title
Warfield,
Robert W.
- III. Petree, Mar-
cella C., jt.
author
- IV. Project

Naval Ordnance Laboratory, White Oak, Md.
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